

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:

Nikolai Mykola Ignatyev et al.

Examiner: Sun Jae Y. Loewe

Serial No.: 10/594,966

Group Art Unit: 1626

Filed: September 29, 2006

For: IONIC LIQUIDS HAVING FLUOROALKYL TRIFLUOROBORATE ANIONS

APPEAL BRIEF

Mail Stop: AF
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

Further to the Notice of Appeal filed on August 12, 2009, please consider the following.

The Appeal Brief fee of \$ 540.00 is filed/paid herewith. The Commissioner is hereby authorized to charge any fees associated with this response or credit any overpayment to Deposit Account No. 13-3402.

(i) REAL PARTY IN INTEREST

The present application is assigned to Merck Patent GmbH, by means of an Assignment filed at Reel 018395, Frame 0624.

(ii) RELATED APPEALS AND INTERFERENCES

There are no related appeals or interferences.

(iii) STATUS OF CLAIMS

Claims 1, 3, 7-12 and 14-17 are pending.

Claims 2, 4-6, 13 and 18 are canceled.

Claims 9-12 and 14-17 are withdrawn.

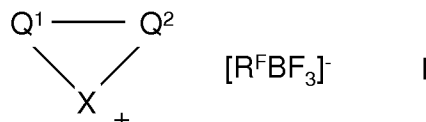
Claims 1, 3 and 7-8 are rejected, and are appealed.

(iv) STATUS OF AMENDMENTS

No amendments to the claims were filed subsequent to Final Rejection.

(v) SUMMARY OF CLAIMED SUBJECT MATTER

The invention is directed to a compound of formula Ia



in which

X denote $\text{N}(\text{R}^1)_2$,

$-\text{Q}^1-\text{Q}^2-$ denotes $-\text{CHR}^3-\text{CHR}^4-\text{CHR}^5-\text{CHR}^6-$,

R^1 in each case, independently of one another, denotes alkyl having 1-10 C atoms or $-(\text{CH}_2)-\text{R}^{11}$,

R^3-R^6 denote hydrogen or alkyl having 1-10 C atoms,

R^{11} denotes perfluorinated or partially fluorinated alkyl having 1-8 and

R^{F} denotes perfluorinated alkyl having 2-8 C atoms.

See the specification at page 3, lines 1-19, and page 4, lines 8-17.

(vi) GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL

The rejection under 35 U.S.C. 103 of claims 1, 3, 7 and 8.

(vii) ARGUMENT

Claims 1, 3, 7 and 8 are rejected under 35 U.S.C. 103 over Zhou taken with MacFarlane. All compounds disclosed in Zhou are imidazolium derivatives. The one paragraph Final Rejection fails to reference, but presumably still relies on, the argument in the Office Action of September 15, 2008 that modification of these compounds to pyrrolidanium compounds would be obvious in view of MacFarlane, as the “disclosed salts have similar thermal behavior such as melting point, viscosity, water miscibility, etc.,” and “one of ordinary skill would be motivated from this disclosure to pair other cations within

their technical grasp (e.g. pyrrolidinium) for the purpose of making additional ionic liquids.” See page 5 of the September 15 Office Action. Appellants disagree with this analysis. In addition to the absence of a close structural similarity, a strong factor suggesting that no presumption of prima facie obviousness exists, one of ordinary skill in the art has absolutely no basis to predict whether the diverse compounds would retain or lose desirable properties with such a significant structural modification to the cation. No rationale is given in the Office Action why one of ordinary skill would pair other structurally diverse cations “within their technical grasp.” Indeed, such random selection of possibilities fails to constitute obviousness, as the Federal Circuit clearly has stated numerous times. For example, in the highly analogous situation in *In re Jones*, 958 F.2d 347, 21 U.S.P.Q. 2d 1941 (Fed. Cir 1992), the Court found that a disclosure in a primary reference of an anion to be used with “ammonium salts” as cations, combined with a secondary reference’s disclosure of a specific ammonium salt, did not constitute a suggestion of the use of that salt of the secondary reference in combination with the anion of the primary reference. Instead, the court held that simply because the cation was a known “ammonium salt” did not suggest its selection from the vast body of such known salts, in the absence of specific direction to do so. Thus, no modification of Zhou is obvious in view of MacFarlane simply because one arguably knows of the MacFarlane cation, in the absence of at least a reasonable expectation of success.

A reasonable expectation of success is not provided by the secondary reference. MacFarlane merely compares imidazolium dicyanamides and pyrrolidinium dicyanamides with respect to their viscosity. The article teaches that 1-ethyl-3-methylimidazolium dca has a viscosity of 21 cP in comparison to N-butyl-N-methylpyrrolidinium dca with 50 cP. This comparison provides no motivation for one of ordinary skill in the art to substitute imidazolium with pyrrolidinium compounds in the primary reference, since viscosity is not alone a determining factor in how the cations might perform as an ionic liquid.

In addition to the lack of structural obviousness and the lack of motivation to make such a significant change as above, the presently claimed compounds are submitted to possess unexpectedly advantageous properties, which clearly eliminate any prima facie case of obviousness. It was clearly not predictable that the electrochemical window of pyrrolidinium pentafluoroethyltrifluoroborate (-3 to +4 as seen from Fig 1 of the present application) would be larger than the disclosed electrochemical window of 1-ethyl-3-methylimidazolium pentafluoroethyl trifluoroborate (-2.5 to + 2.5: Fig. 4 of Zhou et al., page 474). The conclusion of structural obviousness is necessarily predicated on the theory that the cations would have

equivalent effect, which is shown not to be accurate. However, the Office Action of September 15, 2008 argues that these unexpected results are “not statistically significant”, although no reasons for this conclusion have yet been given. Attention is directed to the Declaration Under 37 C.F.R. 1.132 submitted with Appellants’ Reply of January 15, 2009 providing an explanation from an expert in the art why the previous discussion of unexpected results is, in fact, statistically significant. In particular, the Declaration explains that the method used to determine the values has considerable statistical accuracy, so that the improvement shown can be judged as a real improvement.

In addition, the Declaration puts into context the considerable importance of the unexpected increase in stability disclosed from the materials presently claimed. Such increase in stability represents a considerable and needed improvement, as explained in the Declaration, and cannot be ignored.

However, the Final Rejection of May 12, 2009 argues that it is not possible to determine whether the quoted numbers are expected or unexpected, and moreover that it is not persuasive that the unexpected property is sufficient to confer patentability for the instantly claimed compound. Appellants disagree, and attention is directed to the reference of VanSchalkwijk, supplied with Appellants’ Reply of July 10 2009. VanSchalkwijk explains how, in molten salts used in lithium ion and related battery systems, possession of good electrochemical stability is necessary for adequate properties in a battery. See page 187. As noted in section 4.3 on page 199, the electrochemical stability of ionic liquids to anodic oxidation and cathodic reduction, called the “electrochemical window”, is, in the words of the authors, “extremely important for the functioning of battery couples.” As further discussed at page 189, a composition which is less than neutral has a diminished voltage window, i.e., decreased stability and thus utility in batteries. Thus, Figure 2 at page 190 shows that, for a relatively neutral melt, an acceptable voltage window runs from about 2.4 to -2.4. These are similar values as given in the Zhou reference. Note also Figure 11, showing typical stability from about 2 to about -2. Thus, the electrochemical window of the Zhou compositions, -2.5 to +2.5, is typical in the art – and far inferior to the electrochemical window of -3 to +4 shown for the presently claimed materials. It is accordingly evident that, not only is the electrochemical window an important parameter, but the substantial increase in the window with the compositions of the invention, considerably larger than that of the prior art, is not only unexpected but highly advantageous.

However, it is argued in the Advisory Action, in item 11, that the discussion of

VanSchalkwijk is not persuasive, because the reference “discusses the importance of electrochemical stability in the preparation of batteries, not ionic liquids. Therefore it is maintained that the quoted property (i.e. electrochemical stability) is insufficient to confer patentability in Applicant’s invention which is ionic liquids.” This is a misstatement of applicable law. First, it is noted that the electrochemical stability of the ionic liquids is disclosed as being an important parameter thereof, at page 4, lines 19-22 of the specification. Moreover, despite the fact that the specification clearly indicates that electrochemical stability is important for ionic liquids, it is well established that an affidavit directed to an advantage which would “inherently flow” from the normal usage of a claimed material must be considered, even if the specific advantage was not set forth in the specification. See, for example, *In re Zenitz*, 333 F.2d 924, 142 USPQ 158 (CCPA 1964) and *Ex parte Strobel*, 160 USPQ 352 (POBA 1968). In *Zenitz*, patentees claimed pharmacological compound useful as hypotensive agents, antinauseants, antipyretics and sedatives. *Zenitz* submitted affidavits with data alleged to show separation of hypotensive and tranquilizing properties, in other words, that the compounds had either one activity or the other, thus case minimizing side effects if administered either to lower blood pressure or as tranquilizers.. Although the Examiner and the Board of Appeals held that this property, which was not disclosed in the specification, could not be relied upon to show unexpected results, the Federal Circuit’s predecessor court reversed. The court held that the reduced side effects of using a compound in a disclosed pharmaceutical utility would inherently flow from the normal use of the compounds and, thus, must be considered as unexpected results probative of patentability. Analogously, in this present situation, the increased electrochemical stability (which is a property of the ionic liquids, per se) enables highly advantageous use in a significant utility for such ionic liquids, use in batteries. Thus, in accordance with well-established law, the showing of unexpected results in a significant use of the compound must be considered in determination of patentability. The fallacy in the argument of the Advisory Action is apparent when one considers what would happen if the Advisory Action’s analysis were applied to pharmaceuticals. Under this analysis, a pharmaceutical which enabled treating a given disease for which the closest structural analog did not, would not possess unexpected results because the advantage was in a method of using the compound, not in “the compound itself.” Accordingly, it is clear that the data discussed in the specification and declaration must be considered. It is submitted that that data further establishes patentability of the present claims.

In conclusion, it is submitted that ample basis to overturn the rejections of record exists, and the same is respectfully requested.

Respectfully submitted,

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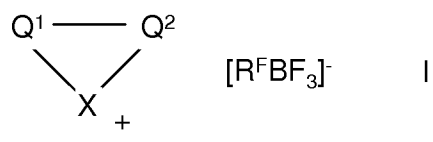
Attorney Docket No.: MERCK-3234

Date: October 13, 2009

HBS/tls

(viii) CLAIMS APPENDIX

1. A compound of the formula Ia



in which

X denote N(R¹)₂,

-Q¹-Q²- denotes -CHR³-CHR⁴-CHR⁵-CHR⁶,

R¹ in each case, independently of one another, denotes alkyl having 1-10 C atoms or -(CH₂)-R¹¹,

R³-R⁶ denote hydrogen or alkyl having 1-10 C atoms,

R¹¹ denotes perfluorinated or partially fluorinated alkyl having 1-8 and

R^F denotes perfluorinated alkyl having 2-8 C atoms.

3. Compounds according to Claim 1, wherein the substituents R¹ are different.

7. A compound according to claim 1, wherein R^F denotes perfluoroethyl, perfluoropropyl or perfluorobutyl.

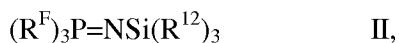
8. A compound according to Claim 1 which is:

N-methyl-N-butylpyrrolidinium pentafluoroethyltrifluoroborate,

N-methyl-N-hexylpyrrolidinium pentafluoroethyltrifluoroborate, or

N-methyl-N-octylpyrrolidinium pentafluoroethyltrifluoroborate.

9. A process for the preparation of a compound according to claim 1,
comprising reacting a compound of the formula II



in which

R^F in each case, independently of one another, denotes perfluorinated alkyl
having 2-8 C atoms, and

R^{12} in each case, independently of one another, denotes alkyl having 1-8 C atoms,
alkoxy having 1-8 C atoms, cycloalkyl having 3-7 C atoms, halogen or aryl,

with a fluoride of the formula III



in which

M is ammonium, alkali metal or alkaline earth metal or a metal from group 11 or
12,

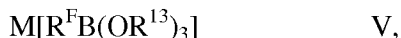
and a boric acid ester of the formula IV



in which

R^{13} in each case, independently of one another, denotes alkyl having 1-8 C atoms
or aryl,

and reacting a resultant salt of formula V



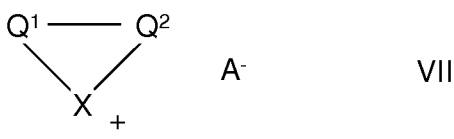
in which M, R^F and R^{13} have one of the above-mentioned meanings,
with HF,

and reacting a resultant salt of formula VI



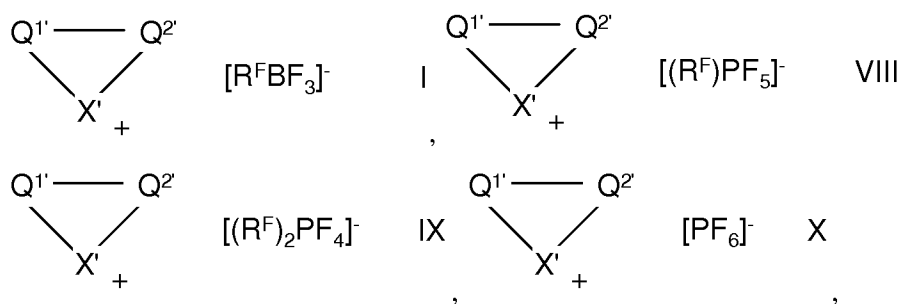
in which R^F is as defined above,

with a compound of the formula VII



in which X and -Q¹-Q²- are as defined for the formula I in Claim 1, and A⁻ denotes alkylsulfate, alkylsulfonate, trifluoromethanesulfonate, tetrafluoroborate, acetate, trifluoroacetate, bis(perfluoroalkyl)phosphinate, F⁻, HF₂⁻, Cl⁻, Br⁻ or I⁻.

10. A mixture of at least one salt of the formula I with at least one salt of the formulae VIII, IX and X.



where

X, -Q¹-Q²- and R^F have the meaning indicated in Claim 1,

X' is NR¹ or N(R¹)₂,

Q¹-Q²- denotes -CHR³-CHR⁴-CHR⁵-CHR⁶,

-CR²=CR³-CR⁴=CR⁵-CR⁶= or

-CR⁷=CR⁸-NR¹⁰-CR⁹=,

R²-R⁶ denote hydrogen or alkyl having 1-10 C atoms,

R⁷-R⁹ denote hydrogen, alkyl having 1-10 C atoms or aryl,

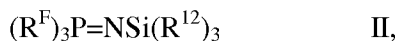
R¹⁰ denotes alkyl having 2-8 C atoms or -(CH₂)-R¹¹, aryl

denotes phenyl, perfluorinated phenyl, or phenyl or perfluorinated phenyl which is substituted by alkyl having 1-8 C atoms.

11. A mixture according to Claim 10, comprising 50-75 mol% of compounds of the formula I and 25-50 mol% of compounds of the formulae VIII, IX and/or X.

12. An ionic liquid containing a compound according to Claim 1.

14. A compound of formula II



in which

R^F in each case, independently of one another, denotes perfluorinated alkyl having 1-8 C atoms, and

R^{12} in each case, independently of one another, denotes alkyl having 1-8 C atoms, alkoxy having 1-8 C atoms, cycloalkyl having 3-7 C atoms, halogen or aryl.

15. A compound according to Claim 14, wherein R^F denotes perfluorinated C_1 - C_4 -alkyl.
16. A compound according to Claim 14, wherein all three substituents R^F are identical.
17. A compound according to Claim 14, wherein R^{12} in each case, independently of one another, denotes alkyl having 1-8 C atoms.

(ix) EVIDENCE APPENDIX

- i. Declaration Under 37 C.F.R. § 1.123 of Nikolai (Mykola) Ignatiyev.
- ii. Article Publication: MATSUMOTO, H., “Electrochemical Windows of Room-Temperature Ionic Liquids,” edited by Hiroyuki Ohno.

(x) RELATED PROCEEDINGS APPENDIX

None